TITLE: C02 Sequestration and Recycle by Photosynthesis

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Abstract

OBJECTIVE

The goal of this project is to gain a fundamental mechanistic understanding of the photocatalytic C02/H2O reaction on semiconductor-based heterogeneous catalysts for the production of hydrocarbons and oxygenates, particularly methane and methanol. This information will enhance the knowledge base in this area and may lead to the design of highly stable catalysts that are active and selective for fuel production under visible light radiation. The following specific tasks have been defined:

- Preparation of a broad range of semiconductor-based catalysts and characterization of their activity and selectivity for the production of methane/methanol from the slurry C02/H2O reaction. This activity and selectivity is examined as a function of reaction conditions, i.e. light intensity, light wavelength, pressure, pH, etc.
- Design and construction of an in situ infrared (IR) reactor that will allow examination of adsorbed species and characterization of active and spectator intermediates.
- Experimentation with use of isotopically labeled species to gain an enhanced understanding of how the reactant fanctionalities are incorporated into the product.
- Application of mechanistic information in conjunction with literature for the design of novel visible light active catalysts.

ACCOMPLISHMENTS TO DATE

Two in situ photocatalytic reactors have been designed and built, one constructed primarily of glass for low-pressure studies and one primarily of stainless steel for high-pressure studies. Both reactors have

been outfitted with IR-transparent windows that allow the IR beam to enter the reactor, pass through the catalyst disk, and exit. In addition, UV and visible light transparent windows have been added at the top of the reactors above the catalyst disk, allowing radiation to pass into the reactor and activate the reaction.
A number of catalysts have been prepared, including Cu/Ti02, Rh/Ti02, Pt/Ti02, Pd/SiC, Cu/SrTi03, dehydroxylated nanoscale Ti02 (nano Ti02), Na2C03/nano Ti02, and layered perovskites- Rb2La2Ti3Olo and K2La2Ti30lO.

The following experiments have been performed thus far, all under ftill spectral UV and visible light radiation and maximum intensity, approximately 250 mW/cm2:

- Slurry C02/1-120 reaction on Cu/Ti02 and Pd/SiC. Cu/Ti02 showed activity for both methane and methanol production while Pd/SiC showed activity for methane only.
- C02/1-120 reaction was studied on Cu/Ti02, Pt/Ti02, nano Ti02, and Na2C03/nano Ti02. Adsorbate formation was observed on Cu/Ti02 and Na2C03/nano Ti02; data is currently undergoing analysis and interpretation. Adsorbate formation was much more facile under conditions in which liquid water was added to the catalyst (T=25'C).

RESEARCH SIGNIFICANCE

C02 emissions are the primary cause of the greenhouse effect. One attractive approach, photocatalytic C02 reduction with water, promises to not only eliminate C02 emissions, but also convert these abundant precursors to transportable fuels. The major challenge to overcome is that the current most active and stable catalysts are only active in the UV region. If a catalyst can be developed that is equally active and stable in the visible light region, the C02/1120 conversion process may become an effective approach to address the potential devastating problem of greenhouse gas emissions.

PLANS FOR THE COMING YEAR

* Propose, develop, and test new catalysts designed for visible light activity.

ARTICLES, PRESENTATIONS, AND STUDENT SUPPORT

Conference Presentations

Scott A. Hedrick and Steven S.C. Chuang, "In Situ Infrared Study of the Photocatalytic Reduction Of C02 with H20 for the Formation of Hydrocarbons and Oxygenates", presented at Pittsburgh-Cleveland Catalysis Society Spring Symposium, May 2000.

Students Supported under this Grant

- Scott A. Hedrick, graduate (Ph.D.) student in chemical engineering, The University of Akron.

- Pisanu Toochinda, graduate (Ph.D.) student in chemical engineering, The University of Akron.
- Dustin Donnelly, undergraduate student in chemical engineering, The University of Akron.